A major purpose of the Technical Information Center is to provide the broadest dissemination possible of information contained in DOE's Research and Development Reports to business, industry, the academic community, and federal, state and local governments.

Although portions of this report are not reproducible, it is being made available in microfiche to facilitate the availability of those parts of the document which are legible.

2

v ,

CONF 870142 --7

LA-UR -87-389

LA-UR--87-389

DE87 006053

TITLE: EXPERIMENTAL SUMMARY

AUTHOR(S): J. L. Smith, Center for Materials Science

SUBMITTED TO: Proceedings of the 5th International Conference on Valence Fluctuations (ICVF), Bangalore, India published by Plenum Press

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

EXPERIMENTAL SUMMARY

J. L. Smith

Los Alamos National Laboratory
Los Alamos, NM 87545, USA

In order to make the experimental summary, I think I must define experiment. Physicists observe things and attempt to understand them. That is probably why most of us chose physics. It so happens that the easy things are all gone. We now go to great lengths to observe things, for example low temperatures, and often to great formalism to understand them, and this has led to a greater distinction between experiment and theory. However our goal is not to work on everything, not to fill tables of data, but to stimulate and enjoy the interplay between experiment and theory. So this is what to look for at a conference.

It seems appropriate to comment first on the name of the conference. As I will mention again there is a continuum in our physics. The topics we discuss are local moment magnetism, heavy fermions, heavy "sodium", even d-electron transition metals, and of course, some mixed valence. I do not necessarily believe that the name of these conferences should be changed, but I would like to recognize the common denominator. Highly correlated electron behavior seems to describe most of the physics we have seen, even if we find the occasional phase transition.

Of course the hottest topic was high-T_c superconductivity in doped La_2CuO_4. Notwithstanding a minority point of view on lanthanum, these new compounds are transition metal materials. Figure 1 shows a Los Alamos measurement of H_{c1} and H_{c2} on one of the superconductors in a squid magnetometer. Varma gave an impromptu overview of what he had seen recently that left us feeling that the superconductivity was genuine. However, I must admit that the talk Rao gave that included all of the metal-insulator transitions in these oxides, which are coupled magnetically, brought back some worry that it may not be superconductivity that has been observed. This issue will certainly be settled soon. It is clear why Fisk said in the panel that we study f electrons to understand transition metals because "that's where the money is." Incidently, the copper in these new superconductors is mixed valent, which is the real reason we were so interested in these materials.

The d-electron transition metals were discussed even more. Riegel can localize 3d's and 4d's by ion implanting them in large lattices. Lonzarich seemed almost embarrassed to speak about systems, such as $TiBe_2$, that have masses ~ 25 m_e and did not show his inelastic neutron data that demonstrate his ideas on overdamped magnetic fluctuations. The data shows

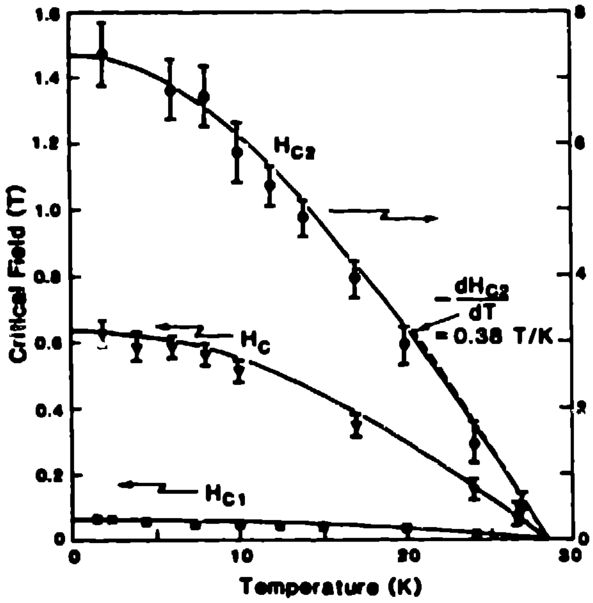

Fig. 1. H_{c1}, H_c, and H_{c2} of $La_{1.7}Ba_{.3}CuO_{4-y}$. This work is submitted for publication by E. Zirngiebl, J. D. Thompson, C. Y. Huang, and C. W. Chu and should be so referenced.

absolutely classical Fermi liquid theory, something that heavy fermions do not.

Two and a half years ago in Köln, Schlabitz gave us superconducting URu_2Si_2 with its spin density wave transition at a higher temperature. Because it has the fourth-highest known electronic heat capacity for a superconductor, we have seen here that this compound is clearly accepted as a full-fledged heavy fermion superconductor along with $CeCu_2Si_2$, UBe_{13}, and UPt_3. Maple showed us that its mass is $\approx 25\ m_e$, the same as Lonzarich's value for $TiBe_2$. Even Wohlleben was moved to give his first talk on heavy fermion superconductors, the point of which was to prove that they were not. At this meeting Schlabitz showed us how to get the T_c up to 1.65 K by adding a pinch of rhodium to URu_2Si_2.

In the $Th_xU_{1-x}Be_{13}$ system, the complete phase diagram remains elusive as I said the first day. Steglich has proposed co-existing, superconducting order parameters in k-space. The only other example of this model is in charge density wave systems. He said that the initial slope of the depression curve must continue on as the second transition line. In the way I plotted this for my talk, I had wondered why it looked this way. Steglich also needs an anomaly at x = 0 as the continuation to x = 0 of the hump in the T_c's. There is the suggestion of this at 0.5 K, as he said, in the heat capacity of pure UBe_{13}, or there is an unpublished, I believe, nagging feature at 0.150 K in the ultrasonic attenuation (Bishop and co-workers). In this same diagram, Maple showed evidence for more than one superconducting phase along the uppermost T_c line based upon both pressure work and the response to gadolinium impurities. He also showed that a critical field scaling that works in one proposed phase does not work in the other. His fits with phenomenlogical models to the critical field curves with gadolinium are quite promising.

Then Heffner ended all of this agreement by vindicating Varma's long-held faith that the second transition was indeed antiferromagnetism. The muon rotation line width increases at the second transition which is only consistent with a one gauss increase in the depolarizing field and is thus very likely due to antiferromagnetic ordering of an effective moment of 10^{-3} μ_B. Anderson pointed out that there remains a slight possibility that it could be consistent with a superconducting state with a net moment. On the whole, little progress was made on describing the superconducting ground states of heavy fermion materials except to note the widespread agreement (Aeppli, Anderson, Asayama, Heffner,...) that they are anisotropic with zeroes on the Fermi surface.

Ytterbium clearly can make heavy fermion compounds. Ott noted that YbP was "a real big one" and that care must be taken to see how γ behaves as temperature approaches zero. Dhar et al. showed $YbPd_2Si_2$ to be heavy and noted that having χ/γ close to one was a crucial point. Rossel and Fisk seconded these ideas.

The problem of the mechanism for how electrons become heavy seems to be yielding to our efforts. Just as Anderson suggested that the four theoretical points of view were converging and that they must all give insight, the experimental work is also converging. Aeppli's accumulated neutron data have shown a common feature of antiferromagnetic fluctuations in all of the heavy electron materials he has studied. Both he and Ott explained how a temperature-dependent RKKY interaction parameter can drive the magnetic order and leave some heaviness behind. Cooper's calculations on orbitally driven magnetism in f-electron materials seem to finally provide a way to deal with the spin-orbit interactions. He can explain the occasional high ordering temperatures in the light rare earths, and also the generally reduced moments and sensitvity to impurities. Lonzarich showed that calculated Fermi surfaces for high mass electrons are quite good and that he could then calculate Ginzburg-Landau parameters to account for the last bit of mass enhancement that is missing from band calculations. His magnetic polaron originating from overdamped magnetic fluctuations seems likely to be the correct physics for the almost magnetic transition metal materials and for the heavy electron materials that have been measured so far by de Haas-van Alphen. Fisk proposed a sort of first cut at a model similar to singlet ground state magnetism where moments can bootstrap themselves as the temperature increases.

As I mentioned earlier, there is no longer any question that there is a continuum in the physics that we have been discussing. It ranges from local f-electron moments through heavy fermions (and the related Kondo problem in dilute systems) through mixed valence to conventional energy band metals with their trivial Fermi surfaces. Because some theorists really do work on heavy sodium, we had a lot of fun talking about spherical cows, which depend on the fact that a cow is topologically equivalent to a sphere which allows us to replace a complex Fermi surface with a sphere of the same volume. Edward's sharp distinction between local and itinerant electrons, which is obvious because one can distinguish whether the electrons contribute to the Fermi surface, occurs between local moments and the heavy electrons, which are just on the edge of being localized. Aside from this, the rest is continuous. Anderson showed deVisser's plot of ln γ versus ln of the density of f centers (a measure of $1/T_K$) to make this point. Fisk outlined a continuum with a very small T_K for local moments. Crow used the systematics of mixtures of $CeIn_3$ (local moment), $CePb_3$ (heavy fermion), and $CeSn_3$ (mixed valent) to show how all of the parameters vary smoothly.

Similarly, Fert, Penney, and Fisk (as would have Moshchalkov if he had come) advocated the Hall effect as an indicator of the onset of coherence (and thus heaviness) and this leads also to an analytic continuation from heavy fermions to mixed valence. Kutty and Vaidya probably saw the same thing under pressure, but must extend their measurements to lower temperature. Croft has compounded cerium with d-electron elements more to the left in the periodic table. In going from chromium to copper he found that cerium exhibited Kondo to mixed valence and back to Kondo behavior. Finally, I believe that Aeppli mentioned that $CePd_3$, a typical mixed valent compound, shows the same sort of spin correlations as the heavy fermion compounds. It was thus made very clear that we must be able to deal with a continuum as we work to understand heavy fermions and mixed valence.

The question of two (or more) temperature scales came up fairly often. I worry about this because the fitting of two exponentials to a curve is not unique. In general one can fit data that looks like the sum of two exponentials with any number of them or even a distribution of exponentials. The fact that such fitting is essentially impossible is not widely appreciated. This can lead people to falsely believe there are two energy scales in their data when other physics is going on. The difficulty is best seen when the two scales then seem to be coupled. Auerbach showed a calculation where such physics can arise from a single energy scale. Thompson, Lawrence, and Wachter all showed work where scaling tends to eliminate the need for two scales or where the use of two is justified by more than one measurement.

The X,1-X business is overworked. I suspect this is done as a safe route to new materials. Most new physics comes from ordered compounds and not from materials of the form $A_xB_{1-x}C_ND_M$. I will give some examples of surprises from ordered compounds. At the Tata Institute: the high ordering temperatures in $CeRh_2Si_2$ and later in $CeRh_3B_2$ have raised many questions as to why there is a departure of ordering temperatures from a DeGennes factor scaling; the mixed valence of $EuPd_2Si_2$ led to much mixed valence work; and dropping a boron into the body center of $CeRh_3$ to yield $CeRh_3B_x$ was a good way to shift properties and is now being put to use in URh_3B_x at Argonne as Dunlap discussed. At Los Alamos, Fisk's pursuit of ordered compounds led him to UPt_3 and its superconductivity, and his attempts at substitutions on one of two different sites in ordered binaries led him to the heaviness of $UAuPt_4$ and $CeAlCu_4$ from the far less interesting compounds UPt_5 and $CeCu_5$. Looking for new ground states in ordered compounds runs the risk, of course, that nothing will be found.

The "sure" route is the X,1-X business, but it usually linearizes the behavior. It guarantees an analytic continuation of the end points with its smoothly varying disorder. The results tend to hide the fact that properties vary logarithmically as impurities are added to the end points. Finally, it makes Anderson tell us, as he did after Ott's talk, that we are putting in too many impurities to be meaningful. I note that the thorium in UBe_{13} and the new high T_c superconductors are X,1-X payoffs. I simply want to suggest that each student not be given another set of end points to linearize.

There were several pieces of work worthy of comment but which do not yet fit into a larger pattern. Boucherle et al. showed that Ce_2Sn_5 had different behavior on its two cerium sites, one local moment and one mixed valent. A beautiful and difficult demonstration. Wachter's structure in the far infrared spectrum of UPt_3 at low temperature must be dealt with. Shivaram showed that an external magnetic field does not orient the superconducting order parameter in UPt_3, again a result of some importance. Rambabu and Malik showed that as germanium replaces silicon in $CeCu_2Si_2$, the p_{eff} is unaffected while the ν moves toward zero to show the magnetic ordering. Grover, Dhar, Umarji, and Malik demonstrated that $CeRh_3B_2$ shows a similarity to heavy fermion materials in its response to impurities. That is, messing with the f-electron sites does not do much, while messing with the other sites has dramatic effects. Edelstein showed that gadolinium impurities in $CeAl_3$ communicate via the cerium atoms. This seems unusual in that local moments do not usually see the heavy electrons.

There were some things that we did not hear about at all. There was no double well discussion and no one lamented that we could not tell itinerant from local f electrons. Indeed, we have made progress over the years. Unfortunately Ron Parks was not here for the fun. He was a man of passion. He loved physics and knew how to instill this in his students. He loved India, after his only visit here, he talked at great length about what he had seen and done. I just heard that he had tried all of the foods from the street vendors without hesitation. Ron's decision to produce the 1969 volumes on Superconductivity was obviously inspired. They remain in heavy use. And of course, he began the mixed valence conferences. He was missed.

Brian Maple and the US-India Cooperative Science Program of the National Science Foundation made possible the fine representation by U.S. Scientists. We are indebted to the students here for all of their efforts to make things go so smoothly. Finally we thank Professors Vijayaraghavan, Gupta, Malik, and many others for their work to make this such an exceptional conference.

This work supported by the National Science Foundation U.S.-India Cooperative Science Program.